THE

SCIENTIFIC PROCEEDINGS

OF THE

ROYAL DUBLIN SOCIETY

VOL. 25 (N.S.).

No. 9.

THE POSSIBLE EXISTENCE OF A FOURTH NATURAL RADIOACTIVE SERIES

BY J. H. J. POOLE, C. F. G. DELANEY AND R. C. MCCORMICK.

DUBLIN :
PUBLISHED BY THE ROYAL DUBLIN SOCIETY.
BALLSBRIDGE, DUBLIN.
HODGES, FIGGIS, AND CO,
6 DAWSON STREET, DUBLIN.
1949.

Price : Three Shillings.

COMMONWEALTH MYCOLOGICAL INSTITUTE
9 FEB 1950

No 9.

THE POSSIBLE EXISTENCE OF A FOURTH NATURAL RADIOACTIVE SERIES.

By

J. H. J. POOLE, C. F. G. DELANEY, AND R. C. McCORMICK.

[Read November 22, 1949. Published January 13, 1950.]

Three radioactive families are known to occur in Nature, i.e. the Th^{232} or 4n, the U^{238} or 4n+2, and the U^{235} or 4n+3 series. The U^{238} and U^{235} series always occur together owing to their parents being isotopic. The heaviest known member of the U^{235} series is Pu^{239}. The half life of Pu^{239} is only 2.4×10^4 yrs., and hence its existence, even at a very early stage of the Earth's history, is improbable, unless it has itself a heavier parent of much greater half life. If Pu^{239} had existed for any appreciable time in the Earth's surface materials, it might have been chemically separated from the U^{238} series, and we might expect to find U^{235} existing independently of U^{238}. One of the few ways of studying radioactivity in past geological ages is by the examination of the pleochroic halos which are produced by small traces of the radioactive elements in certain rock minerals, mainly biotite. Thus uraniun, thorium, and samarium halos have been recognized, but no halo has ever been found that could be attributed to the U^{235} or actinium series. Henderson and Turnbull (1) have however detected the presence of the actinium series in ordinary uranium halos, as would be theoretically expected. Pu^{239} has been found in very small amounts in certain uranium minerals, where it is probably generated by neutron capture by U^{238}.

Recent work, however, in the artificial production of new isotopes of the known radioactive elements has shown that at least six previously unknown radioactive families can be produced. (2 & 3). Of these the most interesting is the Np^{237} or 4n+1 series. The known characteristics of this series are given in Table I.

TABLE I.

NEPTUNIUM OR 4n + 1 SERIES.

Element	Particle Emitted	Half Life	Alpha Ray Energy M.E.V.
$^{94}Pu^{241}$	β	10 years ...	
$^{95}Am^{241}$	α	500 years ...	5.465
$^{93}Np^{237}$	α	2.3×10^6 years ...	4.75
$^{91}Pa^{233}$	β	27 days ...	
$^{92}U^{233}$	α	1.63×10^5 years ...	4.825
$^{90}Th^{229}$	α	7×10^3 years ...	4.93
$^{88}Ra^{225}$	β	14 days ...	
$^{89}Ac^{225}$	α	10 days ...	5.8
$^{87}Fr^{221}$	α	4.8 minutes ...	6.3
$^{85}At^{217}$	α	.018 second ...	7.01
$^{83}Bi^{213}$	β	46 minutes ...	
	$\alpha 2\%$		6.0
$^{84}Po^{213}$	α	4.2×10^{-6} second	8.3
$^{82}Pb^{209}$	β	3.32 hours ...	
$^{83}Bi^{209}$		Stable	

There are several points of interest in these figures. Np^{237} has the longest half life, but as this is only 2.3×10^6 years, it is impossible for the series to still exist on the Earth, unless there exists a heavier member of the family with a half life of at least 500 million years. The first known member of the family is $^{94}Pu^{241}$, and since no beta ray emitter of long life is known to occur among the heavy radioactive elements, it is justifiable to conclude that any possible long-lived progenitor of $^{94}Pu^{241}$ must be an alpha ray emitter, and hence must have an atomic weight of at least 245. No artificial elements of this weight have so far been prepared, so that no direct evidence of their properties can be given. B. Gysae and H. Korsching have published, however, a preliminary note claiming the isolation of a naturally occurring alpha ray emitter with a charge number greater than 92.(4).

It will also be noted that the series contains no noble gas isotope, and that the radium isotope is a beta emitter with a half life of 14 days. In the most generally used methods of measuring the radioactivity of rocks and minerals, it is usually their radon or thoron contents which are determined, and thus such methods would be bound to miss the possible presence of the 4n+1 series. Its presence might however be detected by determining the uranium and thorium contents of the material by the usual methods, and then checking them by counting either the total alpha or gamma emission of the original untreated material.

Experiments on these lines have been carried out by V. H. Hess (5 & 6) and W. D. Urry (7) mainly using Quincy granite. Hess originally found that the gamma ray intensity fron this granite was double that to be expected from its known uranium, thorium, and potassium contents. This result was based on Gray and Tarrant's (8) and Mulhoff's (9) values for the gamma ray emission of potassium. Recently E. Gleditsch and T. Graf (10) have published a new determination of the gamma ray activity of potassium which gives considerably higher values for the number of gamma rays emitted per gramme of potassium. Urry considers that the excess gamma ray emission of Quincy granite is completely accounted for by these results. He has also found that if the uranium, thorium, and potassium contents of the granite are artificially doubled by adding the appropriate amounts of pitchblende, monazite, and potassium carbonate, the gamma ray emission is also doubled. Hess also now holds that his results are satisfactorily explained by the gamma ray emission of the potassium in the granite. The position, however, of the correct values of the radioactive constants of potassium is still rather uncertain (6). It would be interesting to compare the total alpha ray emission of the rock with the calculated figure by the nuclear plate method, since Urry's doubling method does not entirely eliminate the possibility of the gamma ray surplus being due to an unknown radioactive element or family, as the pitchblends or monazite added might also contain the unknown constituent. It would seem very improbable in this case, however, that the gamma emission from the rock should be nearly exactly doubled as Urry found.

The study of pleochroic halos has also given some indecisive evidence on the possible existence of a new radioactive series. Joly (10) in the examination of the black Archaean mica from Ytterby and Arendal found a peculiar type of halo of whose origin he was uncertain. The most striking feature of the halo is a bleached or reversed belt or ring, the bleaching in some specimens being so complete that the original colour of the mica has been destroyed. They differ in appearance from any known type of uranium or thorium halo, but are quite common in the micas named. It seems certain that they cannot be thorium halos, since no trace of a ring large enough to be produced by Th.C′ can ever be detected. Joly, on the whole, was inclined to attribute their origin to an unknown radioactive series.

The simplest method of identifying a fully developed halo is by measurement of its outer diameter. We have re-examined Joly's original material to see if any fresh information on the origin of the halos could be obtained in this way. If the halo was produced by the $4n+1$ series its diameter should be intermediate between those of the uranium and thorium series, the relevant alpha ranges in air being Ra.C′ 6.97 cm., $^{84}Po^{213}$ 7.89 cm., and Th.C′ 8.62 cm. It is clearly impossible to reduce these values exactly to ranges in Ytterby mica, as its stopping power cannot be exactly calculated. Joly estimates that 1 cm. of air is equivalent to between .000436 and .000456 cm. of mica. It was therefore considered that the best method was to determine the average diameter of the uranium halos, which also occur in the mica and compare it directly with the diameter of the unknown or X halos.

The average diameter of the uranium halos was found to be .057 mm. If we assume, as an approximate figure, that the diameter of a halo is proportional to the range of the generating alpha ray, this would mean that the diameter of a $4n+1$ halo should be .0645 mm. The average measured diameter of the X halos was however only .061 mm. It must be pointed out, however, that exact measurements on these halos are very difficult, as the mica itself is very opaque, and the precise position of the edge of the halo largely a matter of the personal opinion of the observer. It seems, however safe to conclude that the difference between .057 mm. and .0645 mm. would have been quite unmistakable, whereas our results show only half the anticipated increase in diameter. It is impossible to distinguish the nucleus in the X halos owing to the complete blackening of the central pupil, and some of their increase in diameter may be due to the nucleus being larger on the average than in the uranium halos. They are also stronger in contrast than the Ra.C′ ring of the uranium halos, and consequently one may be inclined to overestimate their diameter. We would therefore consider, that on the whole, our measurements show that X halos are not produced by the $4n+1$ series. This leaves the problem of their real origin still unsolved.

It might be thought that the radioactivity of the nucleus of a halo could be investigated by the use of nuclear plates. (11 & 12). Unfortunately this method fails owing to the very slow rate of emission of alpha rays from the nuclei of most halos. Joly and Rutherford (13) have estimated the total number of alpha rays required to produce a halo. They used uranium halos in Ballyellen mica from the Mount Leinster Granite, Co. Carlow, Ireland. The age of this granite is probably either late Silurian or early Devonian, that is the halos are about 400 million years old. The number of alpha rays required to produce the darkest halo they observed, is given as 16.5×10^8, or about 4 per year. Experiments on Ballyellen and Ytterby micas with nuclear plates confirmed this result. There was no striking correlation between the alpha ray tracks found in the emulsion and the halos present in the micas. This result would be expected since the exposure times were only about 14 days. The exposure could possibly be increased to two months at most, as after this time the early tracks would tend to become undevelopable. The method therefore was not pursued, as it would seem unlikely to yield any definite results with very old halos, which presumably emit even fewer alpha rays per year than the Ballyellen specimens.

Experiments with nuclear plates on the Ytterby mica, gave nevertheless some indication that it might contain the unknown $4n+1$ series. The average granite emits about 3 alpha rays per hour per cm.^{2} of surface. Mount Leinster mica gave 12 per hour per cm.^{2}, whereas the Ytterby mica gave 800 per hour per cm.^{2}. This corresponds to an uranium content of about 2.5 mg. per gramme, if we attribute all the alpha rays to the uranium family. This is an abnormally high content for any natural material apart from the actual radioactive ores. The result was roughly checked with a Geiger gamma ray counter. A specimen of the mica, weight 190 g., placed near the counter, increased its rate of count from about 38 per min. to 328 per min. Rough tests with cement blocks which contained a known amount of uraninite, showed that the mica should contain about 2.1 mg. uranium

per g. No claim is made for any great accuracy in the counter method, since the apparatus was only designed for detecting fairly high radioactive contents, and would not detect the radioactivity of an ordinary rock specimen. It does show, however, that the radioactivity of Ytterby mica is very high. Swedish Kulm is also known to be very active. It was found that it gave about 130 alpha rays per hour per cm.^{2} by the nuclear plate method, and that 180 g. of it increased the rate of count of the Geiger counter by about 100 per min. showing that the Ytterby mica must be at least three times as active as the Kulm. This fact is perhaps not very surprising, as fergusonite, a rare earth uranium-columbate tantalate, is known to occur at Ytterby. (14). This fergusonite contains from 1 to 2% of UO_2, so that evidently uranuim is fairly plentiful in the district. The alpha rays from the mica are fairly uniformly distributed, and do not show very strong local concentrations, such as are found in many granites. This indicates that the radioactivity of the micas examined was not due to small crystals of fergusonite in them, but that the radioactive elements were moderately uniformly distributed through the material.

Two types of Ytterby mica are known, the unaltered type and the altered or chloritized type. The rare earth elements and the X halos occur in the altered mica. Its radioactivity is also much higher than the unaltered mica. The mica specimens used in these experiments were all of the altered type.

It is theoretically possible to investigage the ratio of uranium to thorium in a flat rock specimen by determining the numerical distribution of the alpha ray ranges observed in a nuclear plate, which has been exposed in contact with the rock surface. (15). If we consider a thick source of a homogeneous single alpha ray emitter, the number of tracks per cm.^{2} in the emulsion, of range greater than r, is given by the expression ;

$$n = \frac{N}{4s} (R - r) \qquad \text{Where,}$$

n = number of tracks with range greater than r (air equivalent) per cm.^{2} of plate
N = total number of alpha rays emitted per c.c. of source
s = stopping power of the source
R = range in air of emitted alpha ray. (16).

$\frac{dn}{dr}$ is therefore equal to $-\frac{N}{4s.}$.

This shows that the number of tracks in the emulsion with ranges between r and $r + \triangle r$ should be constant, and if a histogram of the range distribution is plotted, it should be rectangular in shape. If the source consists of several uniformly distributed alpha ray emitters, a stepped histogram should be obtained.

Lattes, Fowler, and Cuer have investigated the range : energy relationship of the Ilford nuclear emulsion type B1. (17). The manufacturers state that the newer C2 type, which we used, has the same relationship. Table 2 gives the energies and ranges in the emulsion of the known longer range alpha rays as calculated from their figures.

TABLE 2.

Element	Alpha Ray Energy M.E.V.	Range in Emulsion Microns
Th.C′	8.76	47·5
$^{84}Po^{213}$	8.32	43·9
Ra.C′	7.68	38.9
Ac.A	7.40	36.6
$^{85}At^{217}$	7.01	33·7
Actinon	6.82	32.0
Th.A	6.74	31.8
Ac.C′	6.60	30.8
Thoron	6.24	28.4

To use these figures a number *v.* range histogram was made of the tracks in a plate exposed to the Ytterby mica. The horizontal lengths of the tracks were measured with a micrometer eyepiece, and their dips with the vertical slow motion of the microscope. Since an oil immersion objective was employed, and the refractive index of the oil is only about 8% greater than that of the emulsion, the dip as measured may be taken as the actual dip of the track in the emulsion without any large error. This would not be true for a dry objective.

Unfortunately a correction of the measured dip of the track must be made for the decrease in thickness of the emulsion during processing. Presumably the unexposed emulsion is isotropic, and hence the range of the alpha ray would be the same in all directions. The emulsion, however, shrinks to about 60% of its original thickness in processing, and this means that the measured dip will be only 60% of the original dip of the track, and the total range calculated from the horizontal and vertical measurements will be incorrect. In Lattes, Fowler, and Cuer's original determinations the vertical shrinkage of the emulsion introduced no error, since they knew the angle θ (about 4°) at which the particles entered the emulsion, and hence the actual range in the undeveloped emulsion conld be taken as the horizontal length of the track after processing multiplied by sec θ, since the emulsion is prevented from shrinking horizontally by its glass supporting plate. We can approximately correct for the shrinkage of the emulsion by multiplying the measured dips by a factor of 1.7, but in the first histogram made the importance of the shrinkage effect had not been realised, a fact which rather altered its shape.

Fig. 1

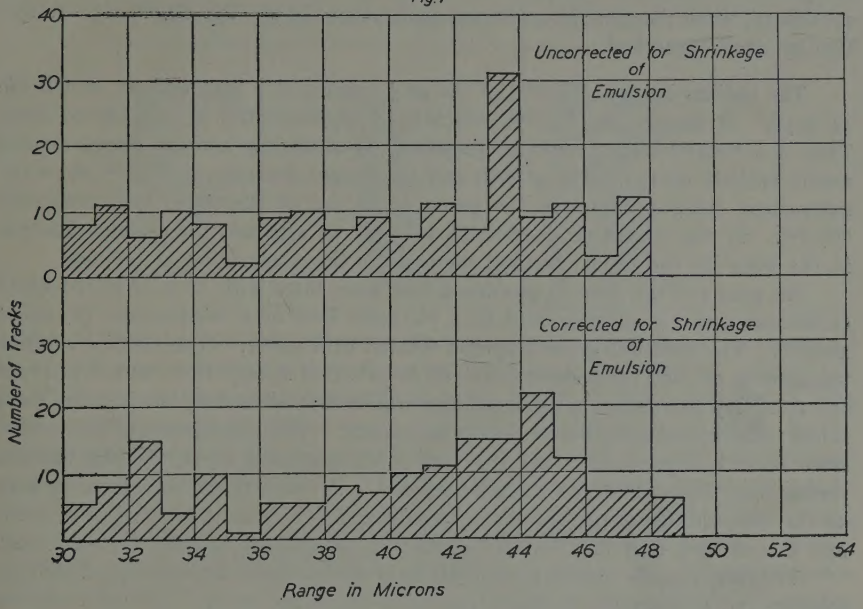


Fig. 1 shows the histogram for the mica uncorrected for shrinkage, and the corrected histogram. In constructing the histograms about 200 tracks were measured. A larger number would have been preferable, but it was thought that 200 tracks was a sufficient number to give a rough idea of the range distribution, and the labour in measuring large numbers of tracks is considerable. It was found unprofitable to measure all the tracks on a given area of the plate, since, as most of them are comparatively short and may originate from many different elements, we can deduce very little from their distribution. Accordingly only tracks, whose horizontal length exceeded 28 microns were measured, and as these were selected by inspection, it is probable that the results are biassed in favour of the longer tracks.

The shape of both histograms is not at all what would be expected from a uniformly distributed thick source, but corresponds more to a surface concentration of radio-activity. The peak in the uncorrected histogram at a range of 43 ot 44 microns is its salient feature. In the corrected histogram this peak is not so pronounced, and has shifted to 44 to 45 microns. It seems probable that the correction for shrinkage has been rather overestimated, as some tracks now have greater lengths than the calculated range of Th.C′ (47.5 microns). The interesting point about this peak is that it corresponds fairly exactly to the range of the alpha ray from $^{84}Po^{213}$, the shortest lived alpha emitter of the $4n+1$ series. This evidence for

the possible existence of the 4n+1 series is undoubtedly very uncertain, but it was considered, when the first histogram was made, that the problem deserved investigation by another method.

The radium isotope, $^{88}Ra^{225}$, in the 4n+1 series, is a beta emitter of half life 14 days. It decays into $^{89}Ac^{225}$, which is an alpha emitter of half life 10 days. Thus if a $^{88}Ra^{225}$ source could be prepared, its alpha ray activity would depend nearly entirely on the initial growth and subsequent decrease of $^{89}Ac^{225}$, since the subsequent decay products in the 4n+1 series are all shortlived compared with $^{89}Ac^{225}$. It was therefore decided to attempt to separate the radium isotopes in the mica by the usual chemical method for the preparation of radium.

Six grams of the mica in powdered form were fused with 18 g. of mixed alkali carbonates and 3 g. of boric acid in a platinum boat at a temperature of about 1000°C. The melt was ground up and treated with water. The alkaline solution was filtered off, and the residue dissolved in 30 c.c. of strong HCl diluted to about 150 c.c. By this means a clear solution was usually obtained, but in some cases it had to be filtered again before further treatment. This procedure is objectionable, since there is a possibility that the very small quantities of the radium isotopes present may be adsorbed on the solid residue. A rough check was therefore kept on the radioactivity of the various rejected products by means of a Geiger counter, and this showed that they had no significant gamma ray activity in most cases.

To precipitate the radium isotopes from the final solution, 0.1 c.c. of 3% $Ba(NO_3)_2$ solution and 10 c.c. of 1.7% Na_2SO_4 solution was added to it. The mica solution was then stirred with a mechanical glass rod stirrer for about 8 hours and the $BaSO_4$ allowed to settle out by standing for a further 18 hours. Most of the supernatant liquid could then be siphoned off, and the $BaSO_4$ precipitate separated from the remainder by filtration through a small $\frac{3}{4}''$ diameter Buchner funnel. The $BaSO_4$ residue was finally washed to remove any acid, which would have affected the nuclear plates subsequently used.

The yield obtained by this method appears to be poor only, at the most about 5% of the expected amount. This may be due to the large volume of the final solution in which the sulphates of the radium isotopes present would be easily soluble. To separate them we are entirely depending on the presence of the $BaSO_4$, which acts as a carrier. We could probably have increased the total yield by using larger amounts of $Ba(NO_3)_2$ and Na_2SO_4, but for alpha ray counts it is preferable to have a small quantity of concentrated active matter, rather than a larger amount of less concentrated material, although the total radioactive content of the latter may be considerably greater. The sources obtained were, however, as active as the alpha ray counter used, which was only a scale of two type, could conveniently handle ; the maximum counting rate being nearly 300 per minute for some sources. The stray alpha ray count was only about 5 per minute, which was a small percentage of the total rate of count for the sources employed.

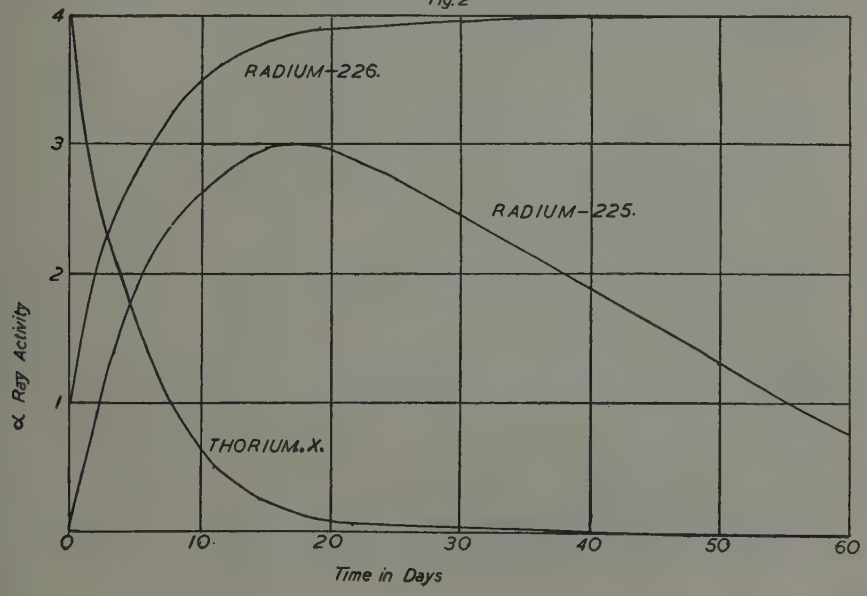
Table 3 gives the radioactive characteristics of the various isotopes of charge number 88, which might be present in these sources.

TABLE 3.

Element	Type of disintegration	Half Life
Ms.Th.1^{228}	beta	6.7 years
Ra^{226}	alpha	1600 years
Ra^{225}	beta	14 days
Th.X^{224}	alpha	3.64 days
Ac.X^{223}	alpha	11.2 days
Ra^{221}	alpha	31 seconds
Ra^{220}	alpha	10^{-2} second

If any of these isotopes are present in the mica solution, they should be separated by precipitation in the same proportions as they have in the solution. Ra^{221} and Ra^{220} have only been produced artificially (3), and it is highly improbable that they could be present in nature, owing to the very short half lifes of all the known members of the radioactive series in which they occur. In any case their presence could not be detected by the method used, since they would break up nearly completely before the source had been prepared. Ms.Th.1 (half life 6.7 years) decays to radiothorium *via* Ms.Th.2, a beta emitter of half life 6.13 hours. Radiothorium is an alpha emitter but since its half life is 1.9 years, its growth in the source will be slow, and consequently the effect of Ms.Th. 1 may be neglected. Similarly, as the activity of Ac.X. will always be small compared to that of Ra^{226}, it may also be neglected.

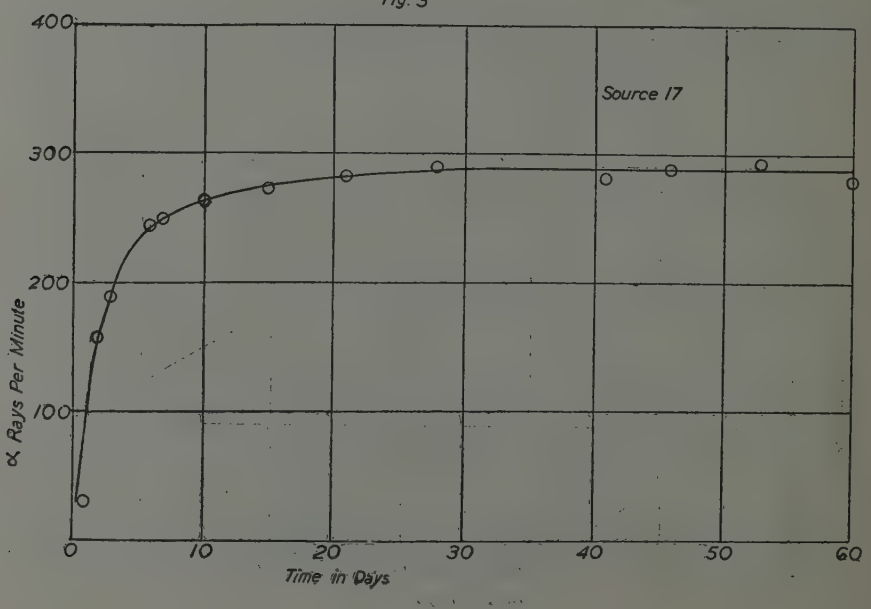
Fig. 2



We may thus consider that the alpha activity of the source obtained from the mica will be mainly due to some mixture of Ra^{226}, Th.X., and possibly Ra^{225}. Fig. 2 shows approximately how the alpha activity of thin sources of the three isotopes would change with time, assuming that none of the disintegration products, such as radon or thoron escape from the source. The maximum activity of the Ra^{225} source is shown as only three quarters of the other two sources. This is done merely for the sake of clarity in the figure. The curve for the surface activity of a thick source of Ra^{226} will not be identical with the thin source curve, but will show a greater ratio between the final and initial activities, due to the longer range alpha rays, produced by the disintegration products of Ra^{226}, escaping from thicker layers than the initial alpha rays from Ra^{226}.

The curves show, however, the fundamental difference in the behaviour of the three isotopes. Th.X. starts with maximum activity and decays nearly exponentially to zero. Ra^{226} shows an initial activity and grows to a practically constant maximum activity of at least four times the initial value in a period of about 30 days. Ra^{225} has zero initial activity, reaches a maximum in 17 days, and falls to half this maximum value in about 47 days. The activity of nearly twenty different sources was investigated, either with an ionisation chamber linear amplifier counter, or by the nuclear plate technique. All sources showed a growth of alpha ray activity with time to a maximum value, which remained constant within the limits of experimental error. Figures 3. and 4. show examples of the type of result obtained by the two methods, Fig. 3. being obtained by the alpha ray counter and

Fig. 3



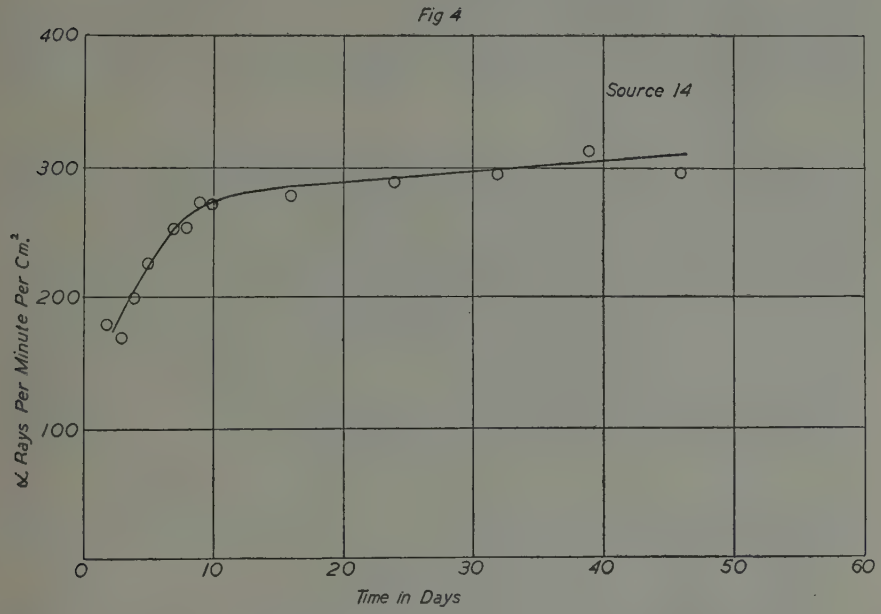


Fig. 4. by the nuclear plate technique. The source used with the alpha ray counter was hermetically sealed by means of a very thin mica window to prevent any loss of radon from the source. Experiments with nuclear plates showed, however, that it was difficult to remove the radon from the sources. A source, placed in a closed vessel which was then evacuated and refilled to atmospheric pressure, this process being repeated several times, showed no significant decrease in activity after treatment. Even heating a source to 800°C for 1 hour *in vacuo*, only reduced its activity to 75% of its original value, whereas if all the radon had been removed and the source considered to be a thin source, its activity should have fallen to about 32% allowing for the growth of radon, etc., during the exposure. The source was over two months old when these trials were made, so that practically no Th.X. could have survived in the source. Thus apparently the loss of radon by the source need not be considered, and the sources, which were used for nuclear plate counts, were usually not hermetically sealed.

For nuclear plate counts only a definite small area of the source was used, and all the tracks produced on the plate by this area were counted. This procedure avoided any error due to lack of uniformity in the distribution of the radioactive material in the source.

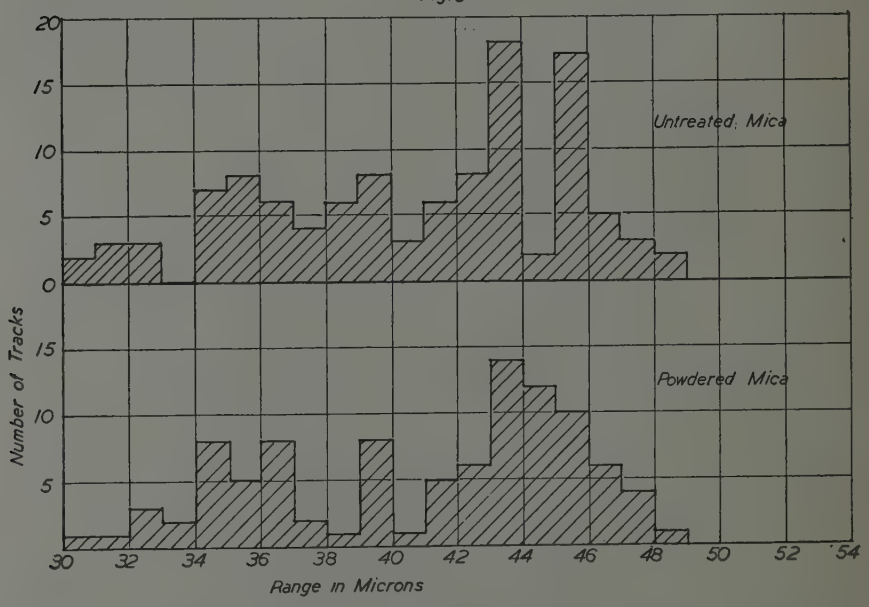
Our results seem therefore to show fairly conclusively that the $4n+1$ series is not present in the mica, at least in amounts comparable with the known natural radioactive series.

The activity curves obtained are in some other respects rather anomalous. From the histogram obtained from the mica surface, it would appear that most

of the radioactivity was due to the thorium series, whereas most of the sources showed a large increase of activity with time, indicating that their activity was mainly due to the uranium series. An attempt was made to clear up this point by some further investigation.

The possibility that the thorium series was being lost in the chemical treatment was investigated by adding a known amount of thorite to the powdered mica. This had the result of increasing the initial activity of the source and decreasing the ratio between the final and initial activities by about the anticipated amount. A Th.X. source prepared from a solution of thorium nitrate of age about 30 years, also behaved normally, falling rapidly with time to a small steady final activity, presumably due to the presence of a small quantity of the uranium family. A new specimen of the mica, whose activity as shown by its gamma ray emission was high, was next more completely examined. Histograms for the untreated surface of the mica and also for the powdered mica were made, about 100 tracks being measured in each case. The results are shown in Fig. 5. Both histograms show

Fig. 5



the usual preponderance of tracks exceeding 40 microns in length, which tracks can only be attributed to Th.C′. The absence of tracks of length 44 to 45 microns in the untreated mica histogram would appear to have no significance, and is probably due to too few tracks being measured. A source was made in the usual manner from the powdered mica, and its alpha ray activity measured with the alpha ray

Fig 6

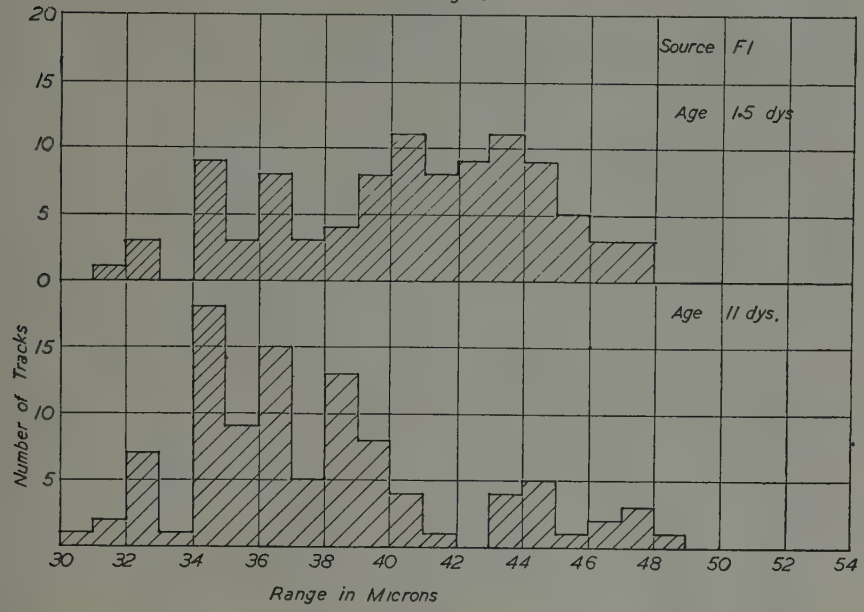
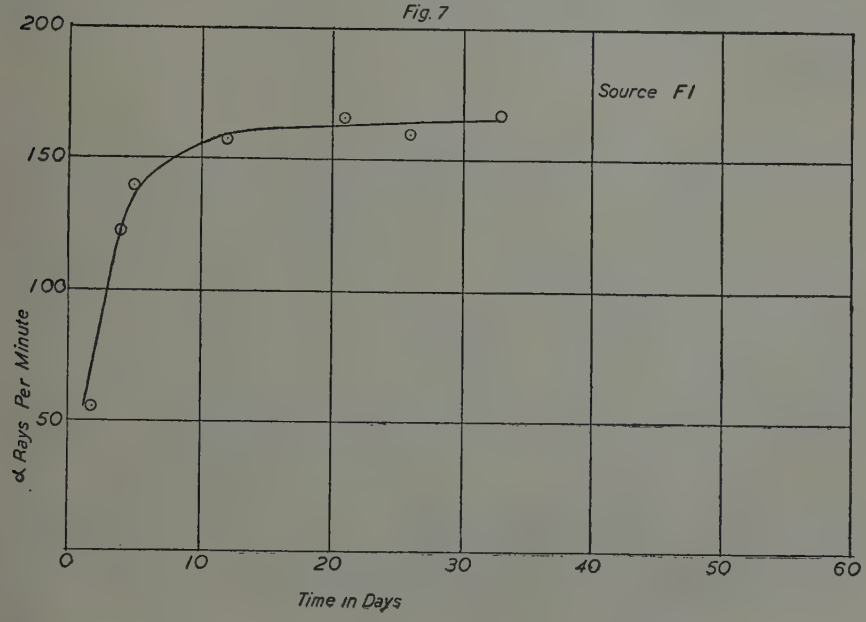

Source F1
Age 1.5 dys
Age 11 dys.
Number of Tracks
Range in Microns
20
15
10
5
0
30
32
34
36
38
40
42
44
46
48
50
52
54

Fig. 7


Source F1
α Rays Per Minute
Time in Days
200
150
100
50
0
10
20
30
40
50
60

counter. The source was left uncovered and exposed to nuclear plates for a period of about 18 hours, firstly at a time approximately $1\frac{1}{2}$ days from precipitation, and secondly at 11 days. Fig. 6 shows the histograms made from the plates, and Fig. 7 the alpha ray activity of the source as determined by the counter method.

The two histograms show clearly the decrease in the proportion of long tracks with time, as would be expected if the long tracks are due to Th.C′. If they had been produced by the 4n+1 series, we would have expected an increase. This again shows the absence of this series in detectible amounts. There is, however, still a discrepancy between the amount of Th.C′. present as indicated by the histograms and the change of alpha ray activity with time as given by the counter. It seems probable that some of this discrepancy is due to the method used in constructing the histograms. The correct method of making them would be to measure the ranges of all the tracks occurring in a certain area of the plate. At first this method was tried, but, as would be expected from a presumably thick source containing both the uranium and thorium series, it was found that only about 1 track in 10 exceeded 30 microns in length. As previously mentioned, this method had to be abandoned, since in order to obtain a histogram of only 100 tracks of range exceeding 30 microns about 1,000 tracks would have to be measured. Since several histograms had to be plotted, the time and labour consumption was considered uneconomical. The tracks of horizontal length exceeding 28 microns were accordingly selected by eye and measured. This procedure undoubtedly favours the longer range tracks since the probability of a track being measured increases with its length, the tendency being to miss the shorter range tracks. It is clear therefore thar undue weight may be given to the long range Th.C′. tracks, as compared with the shorter Ra.C′ tracks.

Recently, however, some peculiar results on the distribution of the radioactivity of certain igneous rocks have been obtained by P. M. Hurley of the Massachusetts Institute of Technology. (18). Granulated samples of these rocks were found to give much higher thick-source alpha ray emissions than predicted on the basis of the radioactive contents of the rocks in bulk. The alpha ray emission was investigated by means of an alpha ray ionisation chamber linear amplifier, which enabled the pulse heights produced by the alpha rays to be determined. In agreement with our results on the Ytterby mica, he found that the distribution of pulse heights suggested a superficial distribution of the radioactive elements on the surfaces of small granules, rather than a uniform volume distribution. He also found that for certain granites the alpha ray activity of the rock powder could be reduced to about 20% of its original value by leaching in hot dilute HCl for about 16 hours. It is intended to apply the same leaching treatment to the mica, since the method offers some interesting possibilities. It is possible, for instance, that for the mica the superficial distribution might be mainly thorium, and the volume distribution mainly uranium. This hypothesis would explain, fairly well, the excess of Th.C′. found by us for the Ytterby mica surface.

SUMMARY.

The general position as regards the possible existence in Nature of the neptunium or $4n + 1$ radioactive series is discussed. The existence in Ytterby mica of unexplained pleochroic halos, discovered by Prof. Joly in 1922, suggests that the neptunium series may be present in the mica. New measurements of the halos showed fairly definitely that these halos are not produced by the neptunium series.

Further experiments on the nature of the radioactivity of Ytterby mica, using both the nuclear plate technique and an ionisation chamber alpha ray counter, are described. The general result of these is to prove that the neptunium or $4n + 1$ series does not exist in the mica, at least in amounts comparable with those of the uranium and thorium series.

The paper concludes with a discussion of some peculiarities observed in the range distribution of the alpha rays emmited by the mica.

Physical Laboratory,
Trinity College,
Dublin.
August 1949.

REFERENCES.

1. G. H. HENDERSON and L. G. TURNBULL.
Proc. Roy. Soc. A. **145**. 582. 1934.
2. G. T. SEABORG.
Chem. Eng. News. **26**. 1902. 1948.
3. W. M. MEINKE, A. GHIORSO and G. T. SEABORG.
Phys. Rev. **75**. 314. Jan. 15. 1949.
4 B. GYSSAE and H. KORSCHING.
Z. Naturforsch. **2***a*. 475. 1947.
5. V. H. HESS.
Phys. Rev. **72**. 609. Oct. 1. 1947.
6. V. H. HESS.
Phys. Rev. **73**. 916. Apr. 15. 1948.
7. W. D. URRY.
Phys. Rev. **73**. 596. Mar. 15. 1948.
8. L. H. GRAY and C. T. P. TARRANT.
Proc. Roy. Soc. A. **143**. 681. 1934.
9. W. MUHLHOFF.
Ann. d. Physik. (5), **7**. 205. 1930.
10. J. JOLY.
Proc. Roy. Soc. A. **102**. 682. 1923.

11. J. H. J. Poole and J. W. Bremner.
Nature **161**. 884. June 5. 1948.
12. J. H. J. Poole and J. W. Bremner.
Nature **163**. 130. Jan. 22. 1949.
13. J. Joly and E. Rutherford.
Phil. Mag. **25**. 644: April 1913.
14. E. S. Dana.
"Descriptive Mineralogy" (6th Edition) p. 729.
15. I. Curie.
J. de Physique. **7**. Serie 8. 313. Nov. 1946.
16. G. D. Finney and R. D. Evans.
Phys. Rev. **48**. 503. Sept. 15. 1935.
17. C. M. G. Lattes, P. H. Fowler, and P. Cuer.
Proc. Phys. Soc. (London) **59**. 883. Sept. 1947.
18. P. M. Hurley.
Unpublished Communication.